Interplay between carrier and impurity concentrations in annealed $Ga_{1-x}Mn_xAs$ intrinsic anomalous Hall Effect

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Investigating the scaling behavior of annealed $Ga_{1-x}Mn_xAs$ anomalous Hall coefficients, we note a universal crossover regime where the scaling behavior changes from quadratic to linear, attributed to the anomalous Hall Effect intrinsic and extrinsic origins, respectively. Furthermore, measured anomalous Hall conductivities when properly scaled by carrier concentration remain constant, equal to theoretically predicated values, spanning nearly a decade in conductivity as well as over 100 K in T_C. Both the qualitative and quantitative agreement confirms the validity of new equations of motion including the Berry phase contributions as well as tunablility of the intrinsic anomalous Hall Effect.

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Rudimentary explanation of the Hall Effect, attributed simply to moving charge carriers experiencing a Lorentz force, 'pressing electricity'[1], spectacularly fails to explain even the simplest of the ferromagnetic materials. From the original measurements of iron foils by E.H. Hall[2] to complex correlated oxide systems[3] to graphene[4], the familiar Hall Effect, from which the nature and amount of carriers can be determined, requires an appellation as 'ordinary' in a sea of extraordinary effects, colorfully termed as 'anomalous' to 'quantum', and even 'extraordinary'. Only recently, has the subtle role of the quantum geometry of the Fermi surface been recognized as intrinsic origins for much of these effects[5, 6]. As any material property determined by transport measurements, the Hall Effect reflects contributions from both intrinsic and extrinsic mechanisms. Separation of the two and the microscopic origins responsible for each has been a source of great contention for decades, especially in magnetic materials, and non-reduced dimensioned materials in general, with limited experimental observations[7, 8] of a clear intrinsic mechanism for the anomalous Hall Effect (AHE).

In the area of semiconductor spintronics, AHE has had an important role in both demonstrating the novelty of carrier mediated ferromagnetic ordering in diluted magnetic semiconductors(DMS)[9] and indirect characterization of magnetic properties[10, 11]. Furthermore, a special case of the AHE with vanishing spin polarization, the intrinsic spin Hall Effect[12], has recently received much interest as possible sources of spins in a spintronic device, utilizing the dissipationless nature of the intrinsic transverse spin current[13]. The idea of dissipationless intrinsic Hall current can be traced to Karplus-Luttinger (KL) formalism[14] in which the term 'anomalous velocity' has been recently reinterpreted as a manifestation of

the Berry curvature of occupied electronic Bloch states. Following KL arguments, the anomalous Hall coefficient (R_S), related to the strength of the spin orbit coupling, scales quadratically with ρ_{xx}, similar behavior to the extrinsic side-jump mechanism in which the carrier is asymmetrically displaced by impurity scattering, proposed by Berger[15]. In between, Smit argued that R_S must vanish in a periodic lattice and proposed the extrinsic skew-scattering mechanism, which predicts the transverse resistivity (ρ_{xy}) to scale linearly with ρ_{xx}[16]. As much experimental evidence showed both linear scaling relationships as well as quadratic, in higher resistive samples, AHE for decades have been thought as an extrinsic phenomenon related to impurity scattering, and KL ideas of intrinsic origins had been discounted. With recent evocation of the Berry phase in the momentum space as the intrinsic origins of AHE, in addition to predicted scaling relationships, measured σ_{xy} can be directly compared to values from well-known material parameters and fundamental quantities to discern intrinsic dissipationless spin currents.

Here, we report a clear and distinct cross-over in the underlying mechanisms for the AHE in a series of annealed $Ga_{1-x}Mn_xAs$. In the intrinsic regime, we observe the transverse conductivity (σ_{xy}) when properly scaled to be independent of longitudinal resistivity (ρ_{xx}), and that the measured values fit recently proposed theories on the intrinsic origins of AHE evoking the Berry phase[6]. Furthermore, the intrinsic dissipationless spin Hall current, impervious to ρ_{xx}, can be manipulated by implicitly and explicitly varying the carrier concentration in $Ga_{1-x}Mn_xAs$.

$Ga_{1-x}Mn_xAs$, a ferromagnetic semiconductor, is one of the most intensively studied materials in the context of semiconductor spintronics, and the recipe for
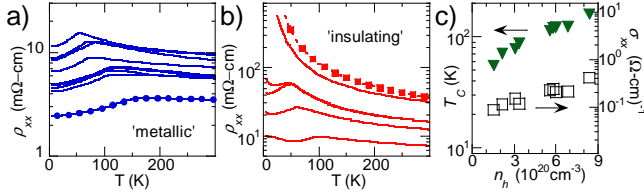
FIG. 1: (a&b) Series of $Ga_{1-x}Mn_xAs$ samples with varying N_{Mn} and N_{Mn}^- that exhibit 'metallic'-like behavior ((a) circle) and that exhibit 'insulating'-like behavior ((b) square) are identified by the sign of $\partial\rho_{xx}/\partial T$ far below T_C. (c) Plot of magnetic (T_C, triangle) and transport (σ_{xx}, open square) properties as function of carrier concentration (n_h).

its growth is well-known[10, 17]. For our study of the AHE in $Ga_{1-x}Mn_xAs$[18, 19], we explicitly vary the total Mn concentration (N_{Mn}) between 2.4 and 6.1% during low-temperature molecular beam epitaxial growth (LT-MBE). Furthermore, we implicitly vary other impurity concentrations by low temperature annealing with temperatures ranging from 200-350 °C. After growth, the samples are fashioned into electrically isolated 300 μm × 1900 μm Hall bar structures. Samples are then annealed in a tube furnace in a flowing dry N_2 environment for one hour with annealing temperature measured by a thermocouple near the sample. After annealing, indium contacts are fashioned and verified as ohmic for transport measurements (Figs. 1&2).

Similar to reports by others[20, 21], we observe the magnetic properties, in terms of the highest magnetic ordering temperatures (T_C), and transport properties, in terms of lowest resistivities, to be optimized at an annealing temperature of ∼250 °C with further deterioration of such properties at higher annealing temperatures. Both our transport and magnetic measurements are consistent with a widely held view that low temperature annealing initially removes Mn_I (N_{Mn}^{++})[22] and other electrically active donor impurities as evidenced by increases in longitudinal conductivity (σ_{xx}), hole carrier concentration (n_h), and T_C. Annealing above optimal temperatures results in deterioration of such properties as $Mn_{Ga}(N_{Mn}^-)$, the source of both spins and carriers, decreases[21]. Even for the highest annealing temperature considered (350 °C), magnetization data along with high resolution x-ray diffraction θ-2θ measurements do not show secondary ferromagnetic phases. However, the existence of such phases cannot be fully ruled out[19]. In short, after LT-MBE and low temperature annealing, a series of $Ga_{1-x}Mn_xAs$ samples exhibiting both insulator- and metallic-like behaviors with σ_{xx} ranging more than two orders of magnitude, n_h ranging from ∼0.7-∼8 × $10^{20}cm^{-3}$, and T_C ranging from ∼25 K to ∼160 K was obtained (Fig. 1.c).

Plotting all the measured ρ_{xx} and ρ_{xy} (Fig. 3), the data indicate an existence of an overall scaling relation-
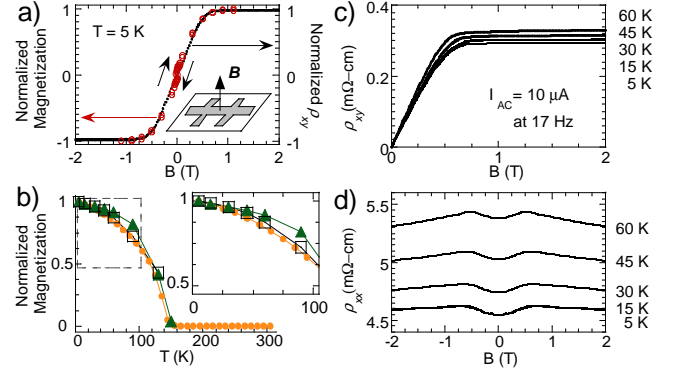

FIG. 2: (a) AHE (closed circle) reflects SQUID magnetometry measurements (open circle) with applied magnetic fields normal to the epifilm for sample $Ga_{0.939}Mn_{0.061}As$ annealed at 250 °C (S6.1-250). (b) Normalized magnetization measurements from SQUID (circle); from Arrott plots of AHE data (triangle); and from $M \propto \rho_{xy}(\partial\rho_{xy}/\partial B)^{-1}$ or $M \propto \rho_{xy}/R_S$ (open square) (S6.1-250). (c&d) ρ_{xx} and ρ_{xy} dependence to applied magnetic field below T_C measured simultaneously by ac lock-in techniques (S6.1-250).

ship akin to the skew scattering origins for the AHE even though ρ_{xx} reflects a wide variance of impurity concentrations. The scaling relationship ($\rho_{xy} = c\rho_{xx}^n$) as applied to $Ga_{1-x}Mn_xAs$ is problematic, as magnetization is inevitably related to ρ_{xx} by n_h as well as weak localization of carriers for higher resistive samples[23]; thus, the relationship $R_S = c\rho_{xx}^n$ may be more appropriate. It has been widely reported that the magnetization of $Ga_{1-x}Mn_xAs$, even for metallic samples, does not easily saturate, thus making a differentiation between the ordinary term and the anomalous term difficult. Thus, we estimate n_h by fitting the temperature dependence of ρ_{xy}[24]. With the anisotropy of the $Ga_{1-x}Mn_xAs$ epilayer in relation to the applied magnetic field and with R_S much larger than the ordinary Hall coefficient (R_o), we determine R_S as $(\partial\rho_{xy}/\partial B)(\mu_o\partial M_Z/\partial B_Z)^{-1}$ from $\rho_{xy} = R_oB + \mu_oR_SM$, an empirical relationship which is upheld for both intrinsic and extrinsic origins of AHE. For the range of x considered in this study, we expect $\partial M_Z/\partial B_Z$ to be constant ($\sim1/\mu_o$) from anisotropic arguments along the lines of Liu et al. and Titova et al.[25]. These two studies find the perpendicular uniaxial anisotropy fields to be nearly independent of n_h and the cubic anisotropy terms to be negligible due to the large built-in strain during LT-MBE. Thus, we estimate cR_S as $\partial\rho_{xy}/\partial B$ (Fig. 2.b).

Plotting $\log(cR_S)$ vs. $\log(\rho_{xx})$ (Fig. 4.a) shows a clear demarcation where a fit to n in the scaling relationship ($R_S = c\rho_{xx}^n$) changes from a value of two to one, for $\rho_{xx} > 10$ mΩ-cm as similarly alluded by Ruzmetov et al.[24], especially at lower temperatures (Fig. 4.b). Here, we vary both temperature and magnetic solute concentrations (N_{Mn}) to vary ρ_{xx}. Furthermore, for temperatures near T_C (Fig. 4.b inset), the scaling relationship seems to
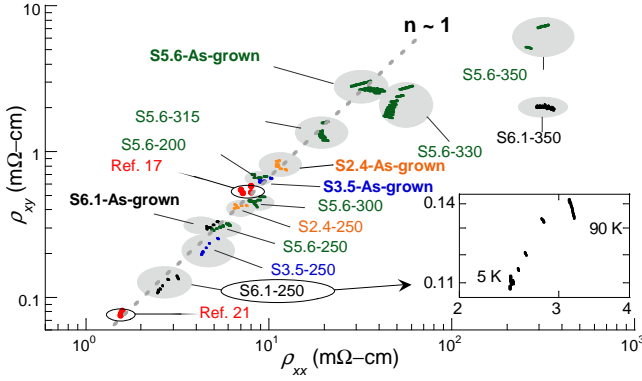
FIG. 3: For each annealed $Ga_{1-x}Mn_xAs$ with differing N_{Mn}^{-} and N_{Mn}^{++}, variations of ρ_{xx} and ρ_{xy} by temperature and applied magnetic fields below respective T_C's are plotted along with data from two other groups (ref. 17 & 21). Each cluster of data points represents an isotherm of a particular annealed $Ga_{1-x}Mn_xAs$ (inset for S6.1-250) ($n \approx 1$ line as a guide).

converge to a value of n equal to one, a result analogous to skew scattering origins of AHE in paramagnetic matrix with embedded magnetic impurities[26], or possibly due to phonon-assisted hopping of holes between localized states in the impurity band[23, 27]. For some of the highest annealing temperatures, we note $n > 3$, suggestive of inhomogeneous systems[19]. Whether the dissipationless anomalous Hall current is intrinsic for all $Ga_{1-x}Mn_xAs$ samples is not clear by examining the scaling relationships alone. Lee *et al.* report of the dissipationless intrinsic AHE in the ferromagnetic spinel $CuCr_2Se_{4-x}Br_x$ maintains that a quadratic relationship between R_S/n_h and $c\rho_{xx}$ should be expected[8]. Interestingly, a similar plot (Fig. 5.a) indicates quadratic relationship between R_S/n_h and ρ_{xx} for all samples considered except for two samples annealed at the highest temperature, 350 °C.

The unexpected quadratic relationship between R_S/n_h and ρ_{xx} for $Ga_{1-x}Mn_xAs$ questioned us what should be the proper normalization method for $Ga_{1-x}Mn_xAs$. The answer comes from the intrinsic AHE theory developed by Jungwirth, Niu and MacDonald (JNM)[6]. In brief, they use a semi-classical transport theory including the effect of Berry phase:

$$\dot{\vec{x}} = \frac{1}{\hbar}\frac{\partial E_n(\vec{k})}{\partial \vec{k}} + \dot{\vec{k}} \times \vec{B}_n(\vec{k}) \qquad (1)$$

$$\hbar\dot{\vec{k}} = -e(\vec{E} + \dot{\vec{x}} \times \vec{B}(\vec{x})) \qquad (2)$$

The resemblance between (Eq. 1) and (Eq. 2) is why the Berry curvature $\vec{B}_n(\vec{k})$ is called "the magnetic field in momentum space". JNM calculated σ_{xy} *exactly* with the assumptions of infinite spin-orbit coupling strength and a mean-field Hamiltonian with the effective field h is given by $N_{Mn}SJ_{pd}$, where N_{Mn} is the density of Mn
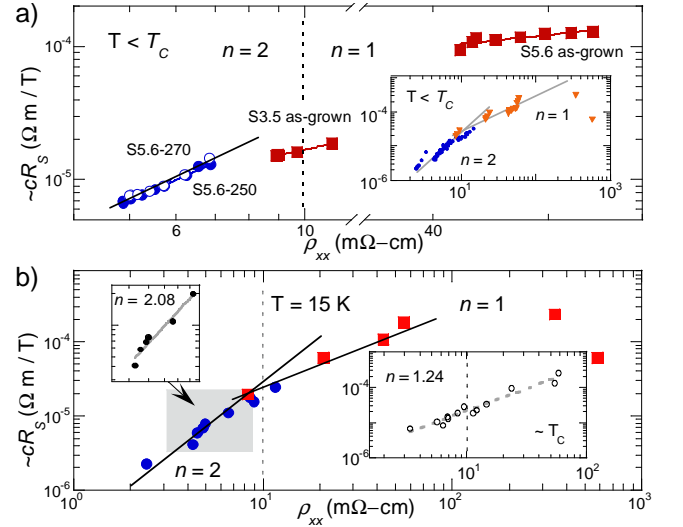

FIG. 4: (a) cR_S ($\propto \partial\rho_{xy}/\partial B$) dependence to temperatures below T_C (each data point represents R_S estimated from AHE measurements at T) and ρ_{xx} are plotted for four different samples: two exhibiting quadratic and two linear behaviors. cR_S for all samples for all measurement temperatures below sample's T_C (inset). (b) For a measurement temperature of 15 K, cR_S dependence to ρ_{xx} is plotted for all samples with differing N_{Mn}^{-} / N_{Mn}^{++}. Lines representing $n = 1$ and $n = 2$ are provided as a guide. Quadratic scaling behavior indicative of intrinsic AHE is found below 10 mΩ-cm and linear above. Plotting similar dependence near T_C for each particular sample show an overall linear-like scaling relationship (inset).

ions with spin $S = 5/2$:

$$CN_{Mn}J_{pd}n_p^{-1/3} < \sigma_{xy} < 2^{2/3}CN_{Mn}J_{pd}n_p^{-1/3} \qquad (3)$$

where $C = \frac{5}{2}\frac{e^2}{2\pi\hbar}\frac{(3\pi^2)^{-1/3}}{2\pi\hbar^2}m_{hh}$. The upper and lower bounds of σ_{xy} correspond to $m_{lh} \approx m_{hh}$ and $m_{lh} << m_{hh}$, respectively, with σ_{xy} of $Ga_{1-x}Mn_xAs$ being closer to the lower bound ($m_{lh}/m_{hh} = 0.16$). In JNM calculations, there exists a tacit condition for a 'clean-limit' where all of N_{Mn} participate in the magnetic ordering as well as later inclusions of finite spin-orbit coupling; warping of band structures; and strains and defects in applying to experimental data[28].

Again, in our scheme to vary ρ_{xx}, we explicitly varied N_{Mn} during LT-MBE growth and implicitly varied N_{Mn}^{++} and N_{Mn}^{-} by low temperature annealing. In course of determining simple scaling relationships of R_S to ρ_{xx}, we observe much of the transport properties and magnetic properties to have a distinct dependence on n_h (Fig. 1.c), as expected from a carrier mediated DMS as $Ga_{1-x}Mn_xAs$[10]. Moriya and Munekata found that N_{Mn}^{-} becomes saturated despite the steady increase of N_{Mn}, and consistent scattering coefficients when N_{Mn}^{-} was used in the room-temperature AHE analysis[29]. Magnetization study also supports this notion: the seemingly deficient magnetization is recovered if only the ionized Mn atoms are counted[30].
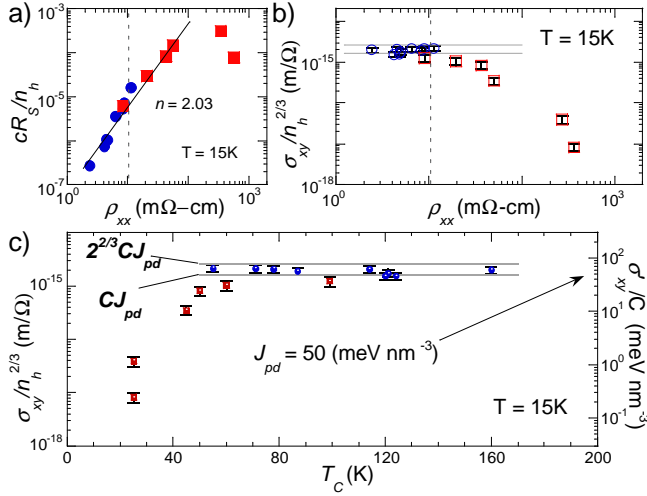
FIG. 5: (a) Normalized cR_S (at T = 15 K) to n_h show a quadratic dependence to ρ_{xx} except for the two highest resistive samples ($n = 2.03$). (b&c) Normalized σ_{xy} (measured at T = 15 K) to $n_h^{2/3}$ for 'metallic'-like samples fits to the 'JNM' lower bound value CJ_{pd} for wide ranging ρ_{xx} (b) and T$_C$'s (c).

Therefore, it is concluded that N_{Mn}^- or n_h rather than N_{Mn} characterizes Ga$_{1-x}$Mn$_x$As epilayers. The monotonic dependences of T_C and σ_{xy} on n_h inferred further support the argument. Then, we find that (Eq. 3) is enough to explain the behavior of metallic Ga$_{1-x}$Mn$_x$As samples once we replace N_{Mn} with n_h. Equation 3 then simplifies to: $\sigma_{xy} = CJ_{pd}n_h^{2/3}$ if we take the lower bound. Thus, we normalize σ_{xy} by $n_h^{2/3}$ instead of n_h and the results are shown (Figs. 5.b&c). Clearly the two classes (metallic and insulating) of Ga$_{1-x}$Mn$_x$As express differing behaviors with the same crossover as the change in scaling behavior of R_S. While the insulating samples show drastic changes as T_C varies, the metallic samples show a good scaling behavior despite the large change in T_C from 50 K to 160 K. The most striking observation is the excellent quantitative agreement with CJ_{pd}, when we use the widely accepted value of $J_{pd} = 50$ meV nm^{-3}.

To summarize, our data clearly shows a universal crossover between intrinsic and extrinsic in the scaling behavior of the anomalous Hall coefficient and that anomalous Hall conductivities of metallic Ga$_{1-x}$Mn$_x$As follows the theoretical prediction qualitatively and quantitatively. And, we find the intrinsic nature of AHE, akin to dissipationless spin Hall currents, in Ga$_{1-x}$Mn$_x$As to be robust.

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